PB87-179248

European Activities in SO2 and NOx Emission Control

(U.S.) Environmental Protection Agency
Research Triangle Park, NC

Apr 87

U.S. Department of Commerce
National Technical Information Service
NTIS

PB87-179248

EPA/600/D-87/117
April 1987

EUROPEAN ACTIVITIES IN SO_2 AND NO_X EMISSION CONTROL

by

William Ellison
Ellison Consultants
Monrovia, MD 21770

Julian W. Jones
Air and Energy Engineering Research Laboratory
U.S. Environmental Protection Agency
Research Triangle Park, NC 27711

EPA Purchase Order 6D1772NASA

EPA Project Officer
Julian W. Jones

AIR AND ENERGY ENGINEERING RESEARCH LABORATORY
OFFICE OF RESEARCH AND DEVELOPMENT
U.S. ENVIRONMENTAL PROTECTION AGENCY
RESEARCH TRIANGLE PARK, NC 27711

REPRODUCED BY
U.S. DEPARTMENT OF COMMERCE
NATIONAL TECHNICAL
INFORMATION SERVICE
SPRINGFIELD, VA. 22161

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO.	2.	3. RECIPIENT'S ACCESSION NO.
EPA/600/D-87/117		

4. TITLE AND SUBTITLE	5. REPORT DATE
European Activities in SO_2 and NO_x Emission Control	April 1987
	6. PERFORMING ORGANIZATION CODE

7. AUTHOR(S)	8. PERFORMING ORGANIZATION REPORT NO.
William Ellison (Ellison Consultants) and Julian W. Jones (EPA/AEERL)	

9. PERFORMING ORGANIZATION NAME AND ADDRESS	10. PROGRAM ELEMENT NO.
Ellison Consultants Monrovia, Maryland 21770	11. CONTRACT/GRANT NO. EPA Purchase Order 6D1772NASA

12. SPONSORING AGENCY NAME AND ADDRESS	13. TYPE OF REPORT AND PERIOD COVERED
EPA, Office of Research and Development Air and Energy Engineering Research Laboratory Research Triangle Park, NC 27711	Published paper; 4/86-3/87
	14. SPONSORING AGENCY CODE EPA/600/13

15. SUPPLEMENTARY NOTES AEERL project officer is Julian W. Jones, Mail Drop 62, 919/541-2489.

16. ABSTRACT The paper gives updated details of major flue gas desulfurization (FGD) and nitrogen oxide (NOx) control installations in the Federal Republic of Germany (West Germany) for coal-fired boilers. The paper reviews applicable government regulations limiting stack emissions, provides an understanding of the principal types of control system designs that have been applied, outlines technological advancements that have been achieved, and reviews experience gained to date in recent expanded use of FGD and NOx control facilities in Europe. Significant differences from FGD service and practice in the U.S. and Japan are described, and specific information is offered that may help improve the design, operation, and reliability of new and retrofit FGD installations in the U.S.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Pollution Emission Sulfur Dioxide Nitrogen Oxides Coal Boilers Regulations Flue Gases Desulfurization	Pollution Control Stationary Sources Europe Japan	13B 05D 14G 21B 07B 07A, 07D 21D 13A

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS (This Report)	21. NO. OF PAGES
Release to Public	Unclassified	13
	20. SECURITY CLASS (This page) Unclassified	22. PRICE

EPA Form 2220-1 (9-73)

NOTICE

This document has been reviewed in accordance with U.S. Environmental Protection Agency policy and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

EUROPEAN ACTIVITIES IN SO_2 AND NO_x EMISSION CONTROL

WILLIAM ELLISON
Director
Ellison Consultants
Monrovia, Maryland 21770

and

JULIAN W. JONES
Senior Chemical Engineer
Air and Energy Engineering Research Laboratory
U.S. Environmental Protection Agency
Research Triangle Park, North Carolina 27711

ABSTRACT

This paper presents updated details of major flue gas desulfurization (FGD) and nitrogen oxide (NO_x) control installations in the Federal Republic of Germany (West Germany) for coal-fired boilers. The paper reviews applicable government regulations limiting stack emissions, provides an understanding of the principal types of control system designs that have been applied, outlines technological advancements that have been achieved, and reviews experience gained to date in recent expanded use of FGD and NO_x control facilities in Europe. Significant differences from FGD service and practice in the U.S. and Japan are described, and specific information is offered that may help improve the design, operation, and reliability of new and retrofit FGD installations in the U.S.

INTRODUCTION

As a result of significant forest damage an unprecedented program of retrofitting of SO_2 and NO_x emission controls on coal-fired boilers is being carried out in West Germany and is leading to substantial steps to achieve reductions in SO_2 and NO_x emissions in other European countries. This major, continent-wide endeavor may influence future programs in North America for control of acid deposition effects.

GOVERNMENT REGULATIONS

West German regulations issued in 1983 and 1984 impose stringent SO_2 and NO_x controls on all boilers above approximately 110 MW(e) and include requirements for retrofitting of FGD on units as small as 35 MW(e). SO_2 and NO_x control compliance is stringently tied to half-hour mean values of emissions (1). This requires gas cleaning systems with design emission levels substantially lower than the regulatory emission-limit values. The 1986 Clean Air Act (TA Luft) (2) imposes stringent SO_2 and NO_x emission limits for comparatively small industrial sources.

SO_2 EMISSION CONTROL

The Emission Control Act (GFAVO) of 1983 (3) requires that coal- or oil-fired units larger than approximately 110 MW(e), comprising 50,000 MW(e) capacity, achieve 85% SO_2 removal, or an SO_2 emission level of 400 mg/Nm^3 (approximately 0.3 $lb/10^6$ Btu), whichever is more stringent, by July 1, 1988. However, based on other provisions of this national directive, boiler owners have designated specific units comprising 12,000 MW(e) for retirement by 1993 after no more than 30,000 additional hours of operation at an SO_2 emission rate not to exceed 2,500 mg/Nm^3 (2.0 $lb/10^6$ Btu). Units in the range of 35 to 110 MW(e) are specified for 60% removal or 2,000 mg/Nm^3 (1.6 $lb/10^6$ Btu) by April 1, 1993. Units from 18 to 35 MW(e) are also required to meet 2000 mg/Nm^3 by April 1, 1993.

NO_x EMISSION CONTROL

A 1984 accord among federal and provincial environmental ministers in West Germany requires that existing coal-fired units larger than 110 MW(e) reduce NO_x emissions to 200 mg NO_2/Nm^3, approximately 100 ppm (vol). Units in the range of 18 to 110 MW(e) are specified for 650 mg/Nm^3 for dry bottom types, and 1,300 mg/Nm^3 for wet bottom (slag tap) types. All existing boilers on a retirement schedule are allowed 650 mg/Nm^3 for dry bottom and 1,300 mg/Nm^3 for wet bottom. Compliance deadlines, set by individual state governments, generally specify 1988 for dry bottom boilers and 1990 for wet bottom boilers. Limits are substantially lower for oil- and gas-fired boilers.

OTHER EUROPEAN LEGISLATION

Influenced by West Germany, Austria, Switzerland, Italy, The Netherlands, Denmark, Sweden, Finland, and the European Economic Community (EEC) have either enacted or proposed significant SO_2 and NO_x control legislation. Table I displays the NO_x emission management objectives of governing officials of the EEC (4), including the 1985 revisions calling for emission rates throughout Europe, after 1995, comparable to current requirements in West Germany.

PRINCIPAL TYPES OF CONTROL SYSTEMS

Wet FGD systems yielding usable by-products and selective catalytic reduction (SCR) NO_x removal systems are the principal technologies being utilized.

SO_2 EMISSION CONTROL

Wet Scrubbing

Lime/limestone scrubbing is the principal FGD method being used. The very large market for this technology (approximately 93% of all capacity sold in West Germany) has been shared by six principal system suppliers operating in West Germany representing a mix of Japanese, U.S., and West German design technologies (5).

TABLE I
EFFECT OF AMENDED EUROPEAN COMMUNITY DIRECTIVE

	Emission Limits (mg NO_2/Nm^3)		
Proposed Directive, 1983	Coal	Oil	Gas
All Plants over 50 MW(t):*			
to 1995:	800**	450	350
after 1995:	400***	220	180
Amended Directive, 1985			
to 1995:			
over 50 MW(t)		450	350
50-100 MW(t)	800**		
100-300 MW(t)	800		
over 300 MW(t)	650		
after 1995:			
over 50 MW(t)		150	100
50-100 MW(t)	400***		
over 100 MW(t)	200		

*100 MW(t) equals approximately 37 MW(e)
**1300 mg/Nm^3 for slag tap (wet bottom) furnaces
***800 mg/Nm^3 for slag tap furnaces

A typical FGD system arrangement that is being extensively applied in European electric-utility applications incorporates the following features similar to those of systems installed in the U.S. in the 1980s:

- Lime or limestone reagent use, with limestone generally preferred.
- Spray-tower-type wet absorber design, rubber-lined.
- In-situ forced oxidation facilities installed in the sump of the absorber without use of sulfuric acid reagent for pH depression/adjustment.
- Primary dewatering of gypsum slurry by liquid cyclones, with final dewatering by centrifuge or horizontal vacuum filter.
- Production of a throwaway or usable grade of gypsum.
- Single-loop wet-scrubbing systems that omit low-pH prescrubbing, used extensively in Japan and in many early West German installations for segregated removal of hydrogen chloride and other minor raw gas components.

Dry Scrubbing

Throughout Western Europe, a considerable number of small and large boilers, including a substantial number in West Germany, are being equipped with conventional throwaway-waste dry scrubbing systems using spray dryers. In instances where suitable solid waste management schemes are available, this technique may be expected to be applied to additional existing West German boilers of 110 MW(e) and smaller by a 1993 FGD start-up deadline. In FGD-using European countries other than West Germany, including Denmark, Austria, and Italy, conventional dry scrubbers are recently being applied at substantially higher inlet flue-gas concentrations (as high as 7000 mg SO_2/Nm^3) than earlier worldwide applications. In some locations, particularly Sweden, spray drying systems are being installed downstream of high-efficiency fly ash collection equipment to more readily permit the FGD solid waste to be converted to a generally usable by-product. In West Germany, commercial circulating-fluid-bed dry scrubbing is being applied, with semidry operation similar to that of spray dryers, at inlet flue-gas SO_2 concentrations as high as 13,000 mg SO_2/Nm^3.

NO_x EMISSION CONTROL

The emission limit (200 mg/Nm^3) set in 1984 for West German boilers larger than 110 MW(e) is well beyond the capabilities of combustion modification techniques and requires the use of flue gas cleaning processes for NO_x removal. Despite the apparent attractiveness of gas cleaning processes that would simultaneously remove both SO_2 and NO_x, and because of imminent compliance deadlines, the commercially developed and applied SCR process (for NO_x removal alone) is being most broadly assessed, adapted, and utilized. This process uses ammonia feed upstream of a catalytic reactor operating at approximately 300 to 400°C (570 to 750°F). A considerable number of selective noncatalytic reduction (SNCR) systems, which operate at 800 to 1000°C (1470 to 1830°F), are also being installed.

COMPARISON WITH THE U.S. AND JAPAN

SO_2 EMISSION CONTROL

As in Japan, most of the West German wet FGD installations are in low/medium sulfur coal service. Similarly, they are designed to employ forced oxidation to yield commercial by-product gypsum and thus avoid the substantial cost and complexity of throwaway management of solid waste. At the same time, application of wet FGD in West Germany differs significantly from that in the U.S. in some major respects:

- With typically low fuel sulfur level (e.g., flue-gas SO_2 concentration of approximately 2000 mg/Nm^3) it has been possible to establish high (95%) availability, system design without use of spare wet scrubber modules.

- o As a nationwide energy conservation measure, FGD installations now under construction have been generally dedicated to use of limestone reagent, and as much as 80% of wet-FGD installation capacity to date uses limestone.
- o Limestone supply is generally ground at the mine source rather than at the boiler site.
- o Due to comparatively high energy cost in Europe, gas is reheated by scavenging heat from the FGD inlet gas. Rotary and tubular regenerative heat exchanger systems are being commonly employed in West Germany (without need for augmented energy supply) to meet a mandatory 72°C (162°F) minimum stack outlet temperature requirement.
- o Due to conservative FGD-buyer policy and the unusual stringency of stack emission control regulations tied to monitoring of half-hour mean values of emission rate, systems are frequently specified by the owner for a substantially lower stack emission rate than the GFAVO emission-limit value.

NO_x EMISSION CONTROL

Approximately 50 SCR pilot plant trains have been installed and tested in West Germany (6) to assess catalyst performance and characteristics. These tests are being performed because of concerns about how applicable the commercial SCR experience on new, base-loaded coal-fired boilers in Japan, albeit extensive, is to existing boilers in West Germany. Most of the concerns are due to the fact that German boilers to be retrofitted with SCR are considered to have a more variable load pattern (more frequent load swings) than those in Japan, which could cause thermal shock of the catalyst structure. The pilot plant testing has included conventional SCR application upstream of the air preheater (high-dust), as well as use of SCR downstream of FGD (cold-side) after flue gas reheating, including 11 with activated carbon. Extensive high-dust SCR field pilot-scale investigation has taken place because possible unique effects of fly ash from German boilers have been of concern. Pilot plant results have indicated that catalyst poisoning is a major problem in application of high-dust SCR to wet-bottom boilers. Because ammonia-slippage problems can lead to fouling of the boiler air preheater, design NO_x removal efficiency substantially above 80% is specified for a number of the German retrofit SCR applications.

DESIGN AND OPERATING EXPERIENCE

SO_2 EMISSION CONTROL

In keeping with the 1988 target compliance-date for SO_2 emission reductions, more than 100 FGD installations, primarily commercial-gypsum-producing types, comprising more than 35,000 MW(e) are being erected. Approximately 50,000 MW(e) of FGD is expected to be in place by the early 1990s. Comparatively few boilers smaller than 110 MW(e) have been retrofitted with FGD to date since their compliance deadline is 1993.

Many FGD facilities already in service in West Germany are the result of a comparatively modest control program that began prior to 1983 under the jurisdiction of individual provincial governments, which led to 20 FGD applications. Thus, although widespread application of FGD in West Germany is comparatively recent, as of the end of 1984 almost 3000 MW(e) of lime/limestone scrubbing systems were in operation, all of single-module slipstream design to meet requirements of pre-1983 legislation. These FGD system installations are reported to have provided acceptable reliability and performance. However, German electric utility specialists note that, with only partial scrubbing of plant flue gas output, these earliest installations have not been subjected to typical full-scale electric-utility-industry operating conditions requiring high availability at sustained high/variable load. Additional FGD capacity of more than 5000 MW(e) originating from the 1983 legislation has now come on-line, but is in an early stage of sustained operation without a lengthy period of experience. However, based on a recent overview (6), system availability of 90 to 95% is being achieved after 12 months of initial operation. Substantial debugging problems/efforts have typically occurred during the initial operations, primarily with gypsum solids processing equipment; i.e., centrifuges, thickeners, filters, dryers, storage facilities, and solids loading and unloading.

Operating with characteristically low or moderate coal sulfur level, and applying commercially demonstrated and conservatively designed forced-oxidation systems, no critical technical problems have been experienced or forecast at the present time. Moreover, because of the comparatively low coal sulfur level, availability concerns have not required the use of spare scrubbing modules. Feasibility of regenerative flue gas reheat using rotary regenerative heat exchangers has been effectively demonstrated. FGD retrofitting has not presented insurmountable problems.

Influenced by this new control activity in Germany, approximately 2000 MW(e) of usable-gypsum-producing FGD systems are being installed in The Netherlands, and a major FGD installation program is underway in Austria on utility boilers. Other European countries, including Sweden, Denmark, Italy, and England, have also become major markets for FGD, with many installations under construction or planned.

NO_x EMISSION CONTROL

More than 10,000 MW(e) of SCR capacity has been purchased for West German plants. The earliest major SCR facility has been installed on a 460 MW(e) bituminous-coal-fired, dry bottom boiler and commenced operation in 1986. Another early installation of 160 MW(e) capacity on a bituminous-coal-fired, wet bottom boiler will achieve close to 90% NO_x removal. Early operations of these facilities are expected to provide important experience on the suitability of very high efficiency SCR installations serving dry- and wet-bottom boilers.

PRINCIPAL TECHNOLOGICAL ADVANCEMENTS

Notable commercial developments in Europe that may be of significant value in stack gas cleaning applications in the U.S. include high sulfur coal applications, regenerative reheat, FGD wastewater treatment, yield of high value gypsum product forms, cold-side SCR, and integrated air-preheater/SCR-reactor design.

HIGH SULFUR COAL APPLICATIONS

European high sulfur coal applications of both the spray dryer and circulating fluid bed extend the electric utility use of these semidry FGD processes to coal sulfur levels substantially above those of present U.S. installations. The circulating fluid bed has had significant European operating experience in this service, and a substantial basis for broader use of semidry scrubbing in the U.S. is developing.

REGENERATIVE REHEAT

Energy economics in West Germany has provided strong motivation for commercial application of regenerative reheat means in high capacity-factor applications. Rotary air heaters, similar to those commonly used for regenerative combustion-air preheating in large boilers, have been extensively used for this purpose with more than 40 purchased, many of which are in operation. Reliable operation of these heat exchangers has followed from application of suitable materials of construction, favorable continuous performance of electrostatic precipitators dedusting the hot inlet gas, and use of high-efficiency, horizontal-gas-flow mist eliminators limiting carryover of FGD scrubbing slurry. On-line, intermittent, high-pressure water washing has effectively prevented exchanger gas pressure drop (the sum of pressure drop across both sides) from exceeding a nominal 125% of the design value for clean conditions. Due to the inherent design of the rotary exchanger, 3-5% of design gas flow leaks from the high-pressure (dirty gas) side to the low-pressure (clean gas) side. Typically, positioning the FGD booster fan on the hot dirty-gas side substantially decreases SO_2 removal. However, a low-leakage design has been developed for this system arrangement that has commercially demonstrated a leakage rate of no more than 0.5% (7).

A comparatively simple, tubular-heat-exchanger type, regenerative reheat system requiring no added flue gas ductwork is also gaining commercial use in Germany after long-term field testing in a lignite FGD installation and may have applicability in the U.S. Non-metallic tubes are exposed to scrubbed gas, and circulating water is the heat transfer medium. Four such systems of an aggregate FGD capacity of 1,200 MW(e) have been installed (8).

At one power plant in West Germany, the wet FGD system is located inside a hyperbolic, natural draft cooling tower serving the plant. The FGD outlet gas intermixes with cooling tower air at a flow rate approximately

25 times greater, at a flow rate ratio (cooling tower air/FGD outlet gas) of about 25:1. Because good atmospheric dispersion is ensured by the discharge and mixing of the FGD exhaust with this substantial air flow, the FGD system is exempted from normal FGD stack exit temperature regulations. For corrosion control a double coating of epoxy is applied to the internal surface of the concrete casing of the cooling tower. The capital cost is reported to be only 20% of that which would be required for a rotary heat exchanger system providing regenerative heating.

FGD WASTEWATER TREATMENT

The application of usable-by-product-forming, wet FGD systems, common in Germany, results in a significant discharge of brackish/saline wastewater. With typically high chloride levels (0.1 to 0.2%) in German bituminous coal, FGD wastewater management is a substantial problem as would be the case at comparable inland sites in the U.S. producing usable by-product. When discharge to receiving streams is not prohibited, the rate of FGD liquor outfall is usually limited to 95 l/m (25 U.S. gpm) per 100 MW(e). When such saline liquor discharge is allowed, trace metal concentrations, dissolved sulfates, and suspended solids must be reduced to specified levels.

Through wastewater purge treatment carried out in the presence of gypsum precipitation from $CaSO_4$-supersaturated liquid, very low trace metal concentrations are achieved prior to outfall discharge (9). A typical wastewater treatment system to accomplish this includes two stages of neutralization, flocculation and sedimentation: the first stage operates at pH 9-10 to remove components such as cadmium and manganese, and the second stage operates at pH 8.5 to remove other components such as zinc and aluminum. An additive in the form of an organic sulfur compound serves as a special precipitation agent to remove mercury. A facility to dewater sludge formed in wastewater treatment is also needed.

YIELD OF HIGH VALUE GYPSUM PRODUCT FORMS

It can be anticipated that West Germany will become a proving ground for gaining the broadest and most diverse use/recovery of FGD by-product solids. Commercially usable FGD gypsum production in West Germany is expected to reach 2.5 million tonnes/year by 1988 and a substantially higher level by 1993. Process innovations are expected to be applied to convert significant amounts of FGD gypsum by-product to the more valuable forms of anhydrite (anhydrous calcium sulfate) and alpha hemihydrate (of calcium sulfate) to help ensure fullest utilization of FGD gypsum by-product. Commercial technology is now available to supply plants to process 100,000 tonnes/year of FGD gypsum yielding highly valued, alpha hemihydrate at a cost as low as 50 DM (approximately $27) per tonne (10).

COLD-SIDE SCR

Many SCR installations that will commence operations by 1990 are being applied downstream of FGD to limit catalyst poisoning, improve unit availability, and, to some extent, because of inadequate plot space to permit normal tie-in of the SCR reactor upstream of the air preheater. This activity in Germany may be expected to provide experience needed to optimize design of SCR facilities for difficult services such as wet-bottom boilers, particularly where it is coordinated with the installation of FGD. In cold-side SCR service, activated carbon (coke) may be of considerable use despite its flammability since its catalysis is effective at a comparatively low temperature [i.e., as low as 90°C (194°F)]; consequently, less reheating of the water-saturated gas is required prior to the SCR step.

INTEGRATED AIR-PREHEATER/SCR-REACTOR DESIGN

Other potentially important developments for NO_x removal include sustained pilot and field test programs in West Germany seeking to adapt existing rotary regenerative air preheaters to incorporate catalyst-coated plates, permitting these facilities to serve also as SCR reactors. Only moderate removal efficiencies are anticipated since catalyst temperature in this economically attractive, integrated arrangement for high-dust service varies within the heat exchanger from 150°C to less than 350°C (302°F to less than 662°F); whereas, an SCR reactor typically positioned upstream of the air preheater ideally operates at approximately 350°C (662°F). Conclusive, large-scale testing work has not yet been reported.

REFERENCES

1. Leutzke, K. and Stahl, H., "Continuous Emission Monitoring for SO_2 and NO_x in the Federal Republic of Germany." Paper presented at the Fourth Seminar on the Control of Sulfur and Nitrogen Oxides from Stationary Sources, Graz, Austria, May 1986.

2. Der Bundesminister des Innern, Umweltschutz, Erste Allgerneine Vervaltungsvorschrift zum Bunbes Immissionsschutzgesetz (Technische Anleitung zur Reinhaltung der Luft-TA Luft) vom, February 27, 1986 (GMBI S 95).

3. Federal Republic of Germany, Thirteenth order implementing the Federal Emission Control Act (large furnaces order) - 13th BImSchV, cabinet decision of June 14, 1983 (German language).

4. Amended proposal for a council directive on limitation of pollutants from large combustion plants. COM (85) 47 final. Brussels: European Commission, February 1985.

5. Der stand der nachruestung bei der rauchgasentschwefelung, elektrizitaetswirtschaft, Jg 85 (1986), Heft 5, pp. 171-174.

6. Forck, B., VGB. Personal communication to W. Ellison, August 26, 1986.

7. Clay, D.E. and Veser, K., Kraftanlagen Heidelberg. Personal communication to W. Ellison, March 25, 1986.

8. Leimkuehler, J., GEA. Personal communication to W. Ellison, January 6, 1986.

9. Forck, B., "Flue Gas Desulfurization and NO_x Reduction." Paper presented at an informal meeting of Swedish electric power companies, Malmo, Sweden, November 6, 1985.

10. Kappe, J. and Ellison, W., "Utilization of Residuals from Flue Gas Desulfurization." Paper presented at the Annual Meeting, American Institute of Chemical Engineers, Chicago, Illinois, November 1985.